Atmospheric
Chemistry
and Physics

Supplement of

Unraveling the role of silicon in atmospheric aerosol secondary formation: a new conservative tracer for aerosol chemistry

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1. Supplementary experimental details

1.1 Sampling of primary source samples of $PM_{2.5}$

For the primary sources of $PM_{2.5}$, seven major sources were collected as literature reported (Ge et al., 2004; Li et al., 2011; Das et al., 2010; Zhao et al., 2006; Chow et al., 2003; Fraser et al., 1998; Hu et al., 2013; Huang et al., 2014; Zhang et al., 2013; Reff et al., 2009; Song et al., 2007; He et al., 2001), including soil dust, industrial emission, coal combustion, vehicle exhaust, biomass burning, urban fugitive dust, and construction dust. The soil dusts were collected at different directions and different depths around Beijing. The industrial emission samples were collected by air sleeves or electrostatic filters installed in the chimneys in steel and power plants in Tangshan, Hebei, which is adjacent to Beijing. The coal combustion samples were fly ashes from burning of honeycomb briquette and lump coals. The fly ashes were drawn through a dilution system (Ge et al., 2004) and then collected onto polypropylene membrane filters by an impactor at a flow rate of 36 L/min. The vehicle exhaust samples were collected following a previously reported method (Fraser et al., 1998) in the middle section of a highway tunnel (Tanyugou Tunnel, Changping, Beijing; 3455 m length) by a high-volume air sampler (Echo Hi-Vol, Milan, Italy) onto Whatman Teflon membrane filters. This tunnel is only open to motor vehicles (mainly diesel- and gasoline-powered vehicles) and the $PM_{2.5}$ concentration outside the tunnel was low (< 15 $\mu g/m^3$), so the collected samples could represent the vehicle emission. The biomass burning samples were collected from burnt straws from Daxing and Tongzhou Districts, Beijing. The urban fugitive dusts were collected in the city of Beijing at an altitude of 20 m by an SYC-3 auto-sampler (Laoshan Instruments, Qingdao, China). The construction dusts were obtained from several construction sites in Beijing and mainly consisted of fine sands, cement dusts and brick dusts.

1.2 Measurement of Si isotopic composition

The Si isotopic composition was measured by multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) on a Nu Plasma II MC-ICP-MS (Wrexham, UK) working in medium-resolution mode coupled to a DSN-100 DeSolvation Nebulizer System. The samples were diluted with HCl solution to obtain a Si concentration of ~300 ng/mL, at which the intensities of ^{28}Si, ^{29}Si and ^{30}Si were about 2, 0.11, and 0.08 V. A PFA nebulizer was used for sample injection at 70 μL/min in dry mode. The optimized instrumental parameters and the Faraday cup configuration are the same as in our previous study (Lu et al., 2018). The procedure blank of the Faraday detector noise was subtracted in the electrostatic analyzer for

30 s prior to each measurement. After each measurement, the system was rinsed with HCl solution (pH 2) for 120 s to reduce the background intensity to < 0.03 V. Three parallel measurements were performed for all experiments (n = 3). The Si isotopic composition was expressed as a δ value (δ^{30}Si and δ^{29}Si) relative to a standard material (NIST SRM-8546) as follows:

$$\delta^{29}\text{Si} = \left(\frac{(^{29}\text{Si}/^{28}\text{Si})_{\text{sample}}}{(^{29}\text{Si}/^{28}\text{Si})_{\text{standard}}} - 1 \right) \times 1000 \qquad \text{(S1)}$$

$$\delta^{30}\text{Si} = \left(\frac{(^{30}\text{Si}/^{28}\text{Si})_{\text{sample}}}{(^{30}\text{Si}/^{28}\text{Si})_{\text{standard}}} - 1 \right) \times 1000 \qquad \text{(S2)}$$

The mass bias of a sample was corrected by using a standard-sample-standard bracketing method based on the mean values of the isotope ratios from two adjacent standard measurements (Lu et al., 2016). Two standard reference materials (NIST SRM-8546 and IRMM-017) were used to validate the method. The δ^{30}Si value of NIST SRM-8546 solution with a Si concentration of 300 ng/mL was measured to be -0.08 ± 0.11‰ (mean ± 2SD, n = 40), and the δ^{30}Si value of IRMM-017 was measured to be -1.38 ± 0.18‰ (mean ± 2SD, n = 36). The δ^{30}Si value of IRMM-017 was highly consistent with the literature reported value (Zambardi and Poitrasson, 2011), confirming the high accuracy and precision of the method.

1.3 Measurement of OC and EC in PM$_{2.5}$

OC and EC were measured by using a DRI Thermal/Optical Carbon Analyzer. The temperature program of the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal evolution protocol was adopted. A punch aliquot of a sample quartz filter was heated stepwise at temperatures of 120 ℃ (OC1), 250 ℃ (OC2), 450 ℃ (OC3), and 550 ℃ (OC4) in a non-oxidizing He atmosphere, and 550 ℃ (EC1) and 700 ℃ (EC2) in an oxidizing atmosphere of 2% oxygen in a balance of He. The carbon that evolved at each temperature was oxidized to CO_2 and then reduced to methane (CH_4) for quantification with a flame ionization detector. The analyzer was calibrated daily with CH_4. Quartz filter blanks were processed concurrently with field blank samples and used to obtain detection limits (LODs). The blank based LODs, calculated as three times the average field blanks, were < 1.0 μg/m^3 for both OC and EC. Replicate analyses were performed at the rate of one per group of 10 samples. The RSDs were less than 5% (Cao et al., 2003). A detailed description of the uncertainties associated with the OC/EC measurements is given in a previous report (Viana et al., 2006).

1.4 Estimation of SOC in PM$_{2.5}$

It has been reported that the minimum value of OC/EC occurred simultaneously in urban and rural areas both in winter and summer (Castro et al., 1999), and particles with the minimum OC/EC ratios were regarded as being dominated by primary aerosols. Thus, the minimum value of OC/EC ratio can be used to estimate the amount of SOC in aerosols and the concentration of SOC can be calculated by the following equation:

$$SOC = OC_{total} - (OC/EC)_{min} \times EC \qquad (S9)$$

where OC_{total} is the total OC, and $(OC/EC)_{min}$ is the estimated minimum OC/EC ratio (Turpin and Huntzicker, 1995).

1.5 Measurement of water soluble inorganic ions in $PM_{2.5}$

The water-soluble inorganic ions were analyzed according to previously reported methods (Tan et al., 2009a; Tan et al., 2009b). The $PM_{2.5}$ filter samples were extracted by an ultrasonication method, and normally > 98% of sulfate, nitrate, and ammonium could be extracted. The filter was submerged in a vial with 10 mL of water, sealed and subjected to ultrasonication for 20 min. The extraction was repeated twice. The extracts were filtered through a 0.45 μm Teflon filter and then analyzed by ion chromatography (Dionex ICS 600 and 2100) to determine the concentrations of water soluble inorganic ions. The concentrations of the water-soluble inorganic ions in the field blanks were 0.22, 0.11, not detected (nd), 0.22, nd, 0.30, nd, 0.36, and 0.19 μg m^{-3} for F^-, Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Ca^{2+}, and Mg^{2+}, respectively. The recoveries for the ions were in the range of 80%-120%. The relative standard deviation (RSD) of each ion was less than 6% in the reproducibility test. Blank values were subtracted from the sample measurements. Specifically, the uncertainties were 4%, 4%, and 5% for SO_4^{2-}, NH_4^+, and NO_3^-, respectively.

1.6 Estimation of secondary aerosols by using the traditional method

Following the traditional method, the secondary aerosol (SA) contribution was estimated by the combination of SIA and SOA. The SIA was the sum of secondary ions (NH_4^+, NO_3^-, and SO_4^{2-}), and the SOA was obtained by multiplying SOC by an OM/OC ratio of 1.6 (Xing et al., 2013).

1.7 Analysis of atmospheric concentration of SO_2 and NO_x and relative humidity

The atmospheric concentrations of SO_2 and NO_x were monitored by a Thermo Scientific Model 43i Sulfur Dioxide Analyzer and a Thermo Scientific Model 42i NO-NO2-NO$_x$ Analyzer, respectively. The relative humidity (RH) was measured by a Vaisala HUMICAP Humidity and Temperature Probe HMP155 (Helsinki, Finland).
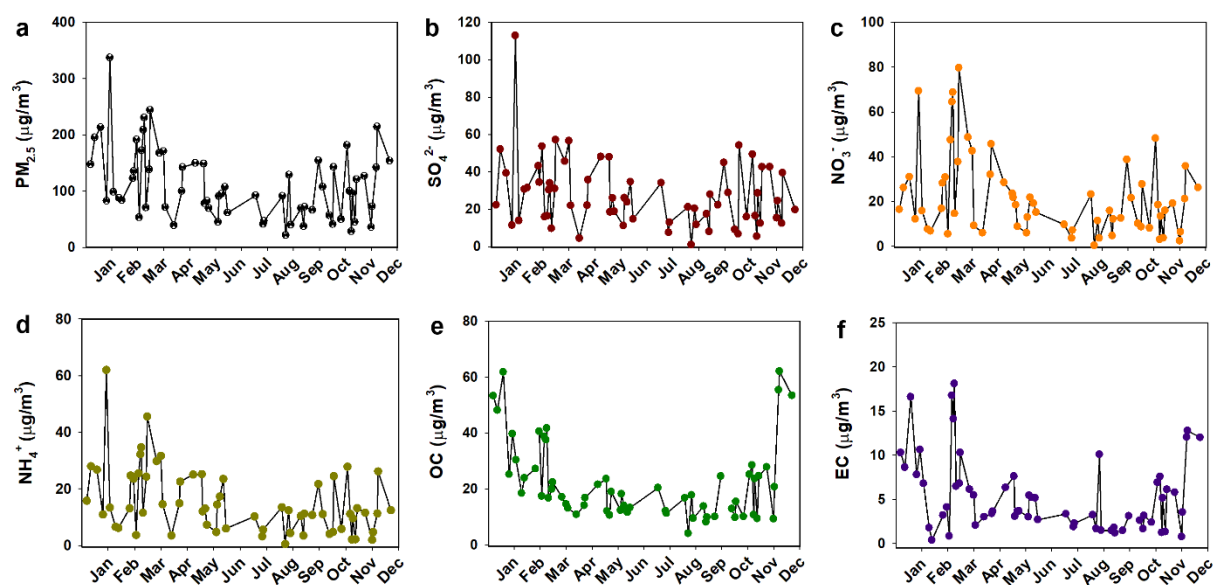
2. Supporting figures

Figure S1. Daily concentration profile of $PM_{2.5}$ (**a**) and secondary species in $PM_{2.5}$ (SO_4^{2-} (**b**), NO_3^- (**c**), NH_4^+ (**d**), OC (**e**), and EC (**f**)) on haze days in 2013. The $PM_{2.5}$ samples were collected around Beijing on haze days in 2013 (see the Sect. 2.2 in the paper) (Lu et al., 2018). From (**a**), the $PM_{2.5}$ concentration showed large variations among different days ranging from 28.4 to 337.1 $μg/m^3$ with an annual mean value of 112.4 $μg/m^3$. The pollution level in spring and winter was higher than that in summer and autumn probably due to the larger emission of coal combustion in spring and winter in this region (Lu et al., 2018; Liu et al., 2016). From (**b**)-(**f**), all secondary species in $PM_{2.5}$ showed a similar seasonal trend with the $PM_{2.5}$ (i.e., higher in spring/winter than in summer/autumn), because secondary aerosols are a major component of $PM_{2.5}$ in Beijing (Huang et al., 2014). While for individual days, the concentrations of different secondary species showed large differences, suggesting different sources and secondary particle contribution to $PM_{2.5}$ on different haze days.
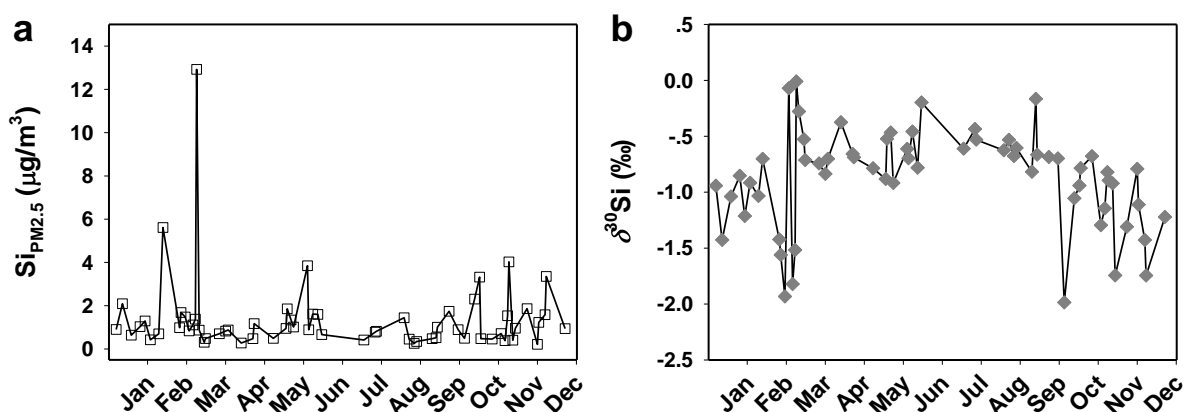
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Figure S2. The daily mean $Si_{PM2.5}$ (**a**) and Si isotopic composition ($\delta^{30}Si$) of $PM_{2.5}$ (**b**) on different haze days in 2013. The sample information was the same as in Fig. S1. From (**a**), the $Si_{PM2.5}$ did not show any seasonal trend, which was totally different from the concentration profiles of $PM_{2.5}$ and secondary species in $PM_{2.5}$ (as shown in Fig. S1). However, for $\delta^{30}Si$ of $PM_{2.5}$ (**b**), a clear seasonal trend that more negative in spring/winter than in summer/autumn was observed. It should be noted that the Si isotopic signature could indicate the primary sources of $PM_{2.5}$ (Lu et al., 2018). Thus, the results in (**b**) suggested that [30]Si-depleted sources (e.g., coal combustion and industrial emission (Lu et al., 2018)) contributed more to aerosols in spring/winter than in summer/autumn. The discussion above evidenced that the airborne Si in $PM_{2.5}$ was only related to the primary source emission but did not depend on the aerosol formation.

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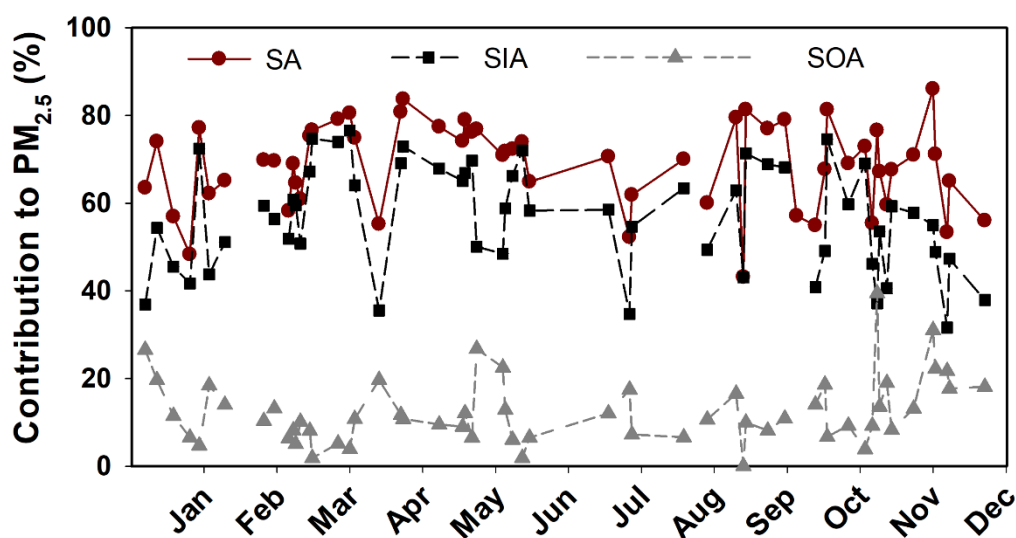
Figure S3. Contribution of secondary aerosol (SA), secondary inorganic aerosol (SIA), and secondary organic aerosol (SOA) to $PM_{2.5}$ estimated by using the traditional method. The contributions were estimated based on the chemical compositions of $PM_{2.5}$ (e.g., SO_4^{2-}, NO_3^-, NH_4^+, OC, and EC). It can be seen that the SA contribution was quite high all through the year, and the contribution of SIA was much higher than that of SOA.
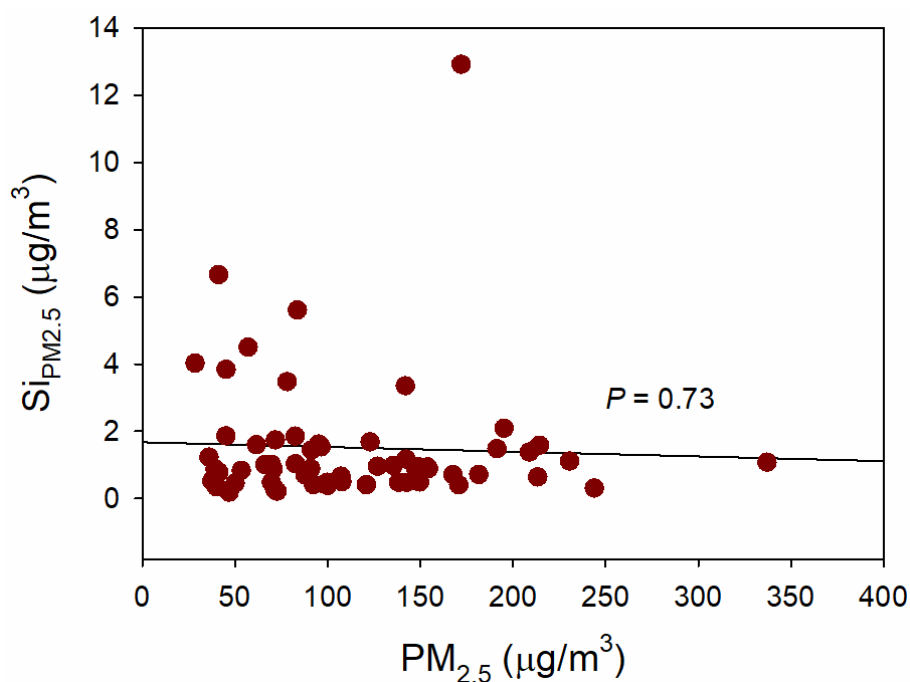
Figure S4. Correlation analysis of total Si in $PM_{2.5}$ ($Si_{PM2.5}$) with $PM_{2.5}$ concentration on haze days. The Si in $PM_{2.5}$ did not show any correlation with the $PM_{2.5}$ concentration ($P = 0.73$). With the increase of $PM_{2.5}$ concentration, the $Si_{PM2.5}$ was unaffected and kept a steady trend. Considering the high secondary aerosol contribution to $PM_{2.5}$ in the studied region, this results also suggested that the airborne Si might not be involved in the secondary aerosol formation.
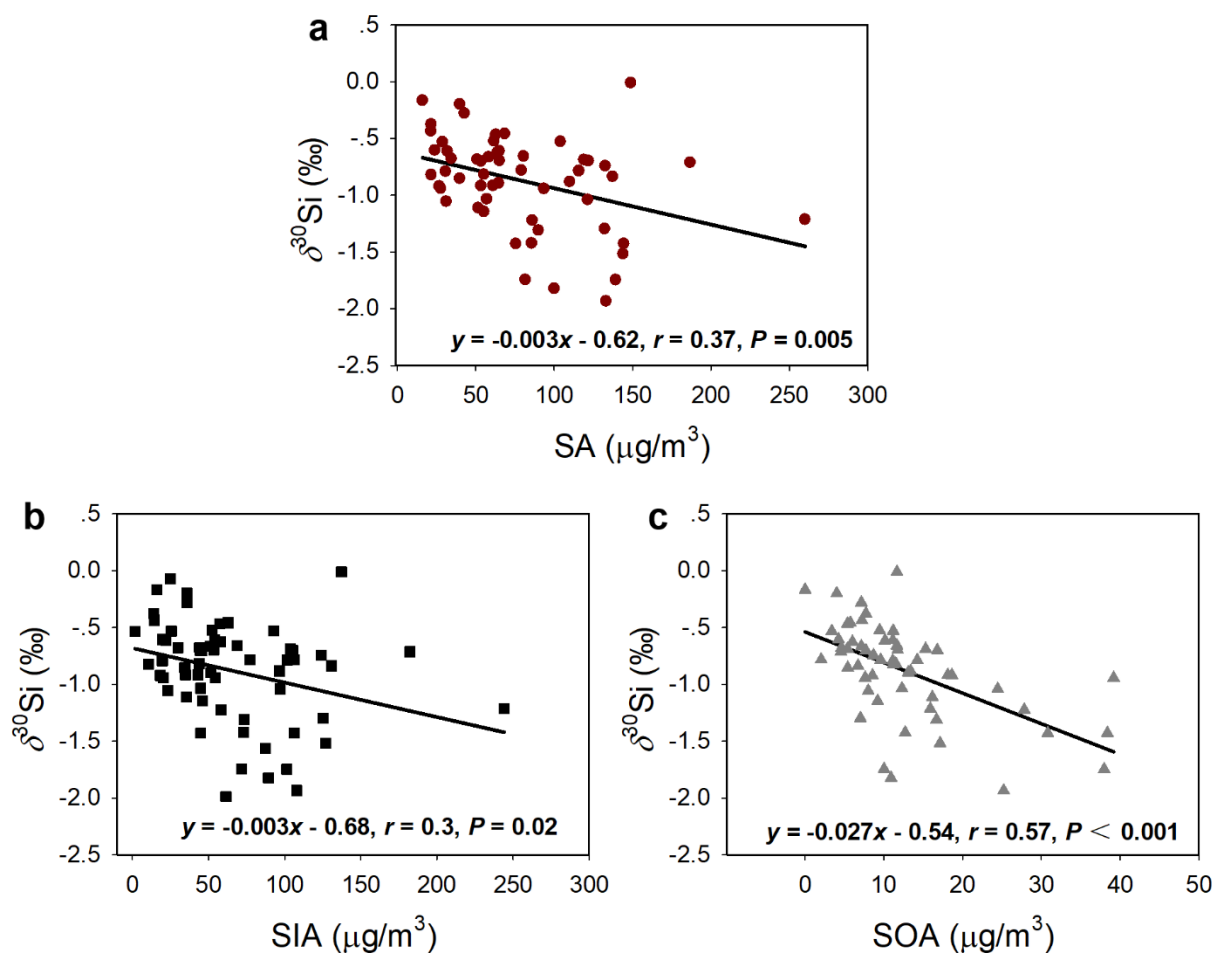
Figure S5. Correlation analysis of Si isotopic composition (δ^{30}Si) of PM$_{2.5}$ with the secondary aerosols (SA (**a**), SIA (**b**), and SOA (**c**)).
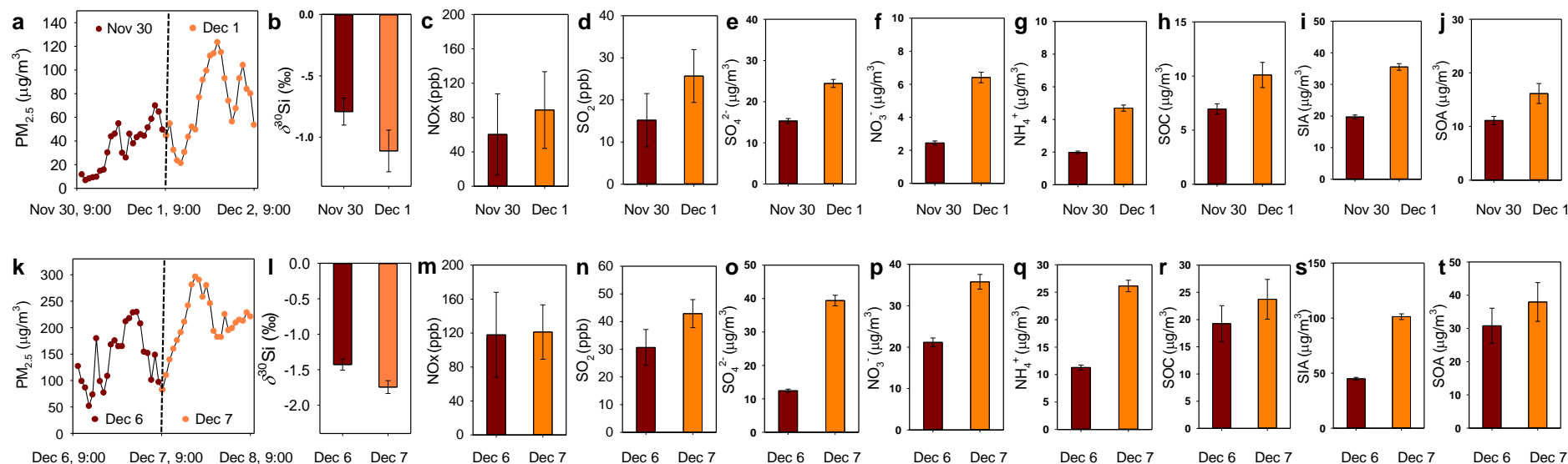
Figure S6. Variations of Si isotopic composition ($\delta^{30}Si$) and multi-chemical species of secondary aerosols (NO_x, SO_2, SO_4^{2-}, NO_3^-, NH_4^+, SOC, SIA, and SOA) during two typical haze events. (**a**), Monitoring of $PM_{2.5}$ concentration during Nov 30, 2013 to Dec 1, 2013. (**b**), Daily Si isotopic composition ($\delta^{30}Si$) of $PM_{2.5}$ in **a**. (**c-d**), Concentrations of precursor NO_x (**c**) and SO_2 (**d**) of SA in **a**. (**e-j**), Concentrations of secondary species SO_4^{2-} (**e**), $NO_3^-$ (**f**), NH_4^+ (**g**), SOC (**h**), SIA (**i**), and SOA (**j**) of SA in **a**. The red and orange bars in (**b-j**) correspond to the days marked with the same colors in **a**. (**k**), Monitoring of $PM_{2.5}$ concentrations during Dec 6, 2013 to Dec 7, 2013. (**l**), Daily $\delta^{30}Si$ of $PM_{2.5}$ in **k**. (**m-n**), Concentrations of precursor NO_x (**c**) and SO_2 (**d**) of SA in **k**. (**o-t**), Concentrations of secondary species SO_4^{2-} (**o**), NO_3^- (**p**), NH_4^+ (**q**), SOC (**r**), SIA (**s**), and SOA (**t**) of SA in **k**. The red and orange bars in (**l-t**) correspond to the days marked with the same colors in **k**. The error bars represent 1SD from three parallel Si isotope measurements in (**b**) and (**l**), the concentrations of SO_4^{2-} (**o**), NO_3^- (**p**), NH_4^+ (**q**), daily variation of NO_x (**m**) and SO_2 (**n**), and uncertainties of calculation on the concentrations of SOC (**r**), SIA (**s**), and SOA (**t**).

3. Supporting tables

Table S1. Concentrations of PM$_{2.5}$ and meteorological parameters in the sampling days.[a]

Date	PM$_{2.5}$ (μg m^{-3})[b]	T_a (℃)	RH (%)	P_{vapor} (hPa)	WS (m s^{-1})	WD (Degree)	P (hPa)	T_g (℃)	R_{total} (W m^{-2})	PAR (μmol s^{-1}m^2)
6/1/2013	147.4±24.5	-4.4	30.9	1.3	1.0	145.6	1020.3	-4.3	79.6	113.5
11/1/2013	195.1±24.0	-2.7	55.8	2.7	1.2	276.9	1016.7	-2.0	57.5	81.7
18/1/2013	213.4±15.3	-2.0	43.3	2.2	0.8	172.0	1019.9	-0.9	84.6	114.0
25/1/2013	82.6±29.4	-4.2	32.5	1.5	1.2	93.0	1016.4	-3.0	89.6	120.7
29/1/2013	337.1±9.9	-2.3	80.8	4.2	0.9	245.5	1017.9	-0.1	33.5	50.6
2/2/2013	98.3±11.3	-1.2	32.2	1.8	1.4	126.4	1023.9	1.3	109.4	146.2
8/2/2013	87.7±7.7	-6.9	29.6	1.1	1.6	141.5	1028.1	-4.8	122.4	157.2
11/2/2013	83.6±31.1	-3.7	34.4	1.7	1.4	132.8	1025.2	-1.5	66.5	98.6
23/2/2013	122.8±8.0	4.0	24.0	1.9	0.8	171.5	1020.0	9.2	166.5	266.2
24/2/2013	135.4±22.9	3.1	33.2	2.5	0.9	177.7	1019.3	7.4	102.3	165.6
27/2/2013	191.3±15.8	7.4	40.8	3.5	1.6	207.9	1009.4	12.3	171.9	275.0
2/3/2013	53.4±8.8	1.0	21.5	1.4	1.0	125.3	1025.4	4.8	76.5	138.9
5/3/2013	172.0±6.8	8.2	26.8	2.9	0.8	170.8	1009.4	13.1	124.2	195.0
6/3/2013	208.8±10.7	11.4	35.9	4.5	1.1	161.1	1000.9	16.1	141.2	226.2
7/3/2013	230.5±7.3	11.9	36.6	4.9	1.4	135.8	997.7	16.7	134.3	211.4
10/3/2013	70.5±4.5	7.0	15.5	1.6	2.4	186.5	1018.7	11.6	210.0	322.1
14/3/2013	138.2±7.0	4.4	50.4	4.1	1.8	189.8	1019.9	9.4	161.6	217.3
15/3/2013	243.9±2.9	ND	ND	ND	ND	ND	ND	ND	ND	ND
26/3/2013	167.6±5.4	8.0	43.1	4.6	1.2	244.6	1006.0	13.3	167.0	238.2

31/3/2013	170.7±4.2	5.3	74.8	6.7	1.9	154.8	1011.6	9.3	33.1	55.2
2/4/2013	71.4±4.8	11.7	31.6	4.2	1.8	205.6	1010.0	17.4	225.5	334.7
12/4/2013	39.2±4.8	15.6	19.5	3.5	1.8	191.3	1005.3	21.7	231.6	355.1
21/4/2013	99.8±7.3	12.5	45.6	6.4	2.0	209.8	1010.7	18.3	226.7	328.7
22/4/2013	142.3±9.6	12.7	57.3	8.5	1.7	170.4	1006.6	17.5	111.7	171.1
6/5/2013	149.6±8.9	ND	ND	ND	ND	ND	ND	ND	ND	ND
7/5/2013	148.5±8.4	24.3	39.4	11.7	1.1	199.6	986.5	29.0	173.7	261.5
17/5/2013	78.1±5.9	21.4	50.5	12.8	1.2	182.8	1004.2	25.2	124.4	196.8
18/5/2013	82.5±3.4	21.3	59.4	14.9	1.5	182.3	998.8	24.2	92.1	138.8
21/5/2013	69.6±11.7	23.4	35.8	10.3	1.2	158.0	991.9	27.3	193.8	298.3
23/5/2013	45.1±6.3	24.4	29.8	9.0	0.9	189.6	1002.8	29.0	211.1	325.0
3/6/2013	90.8±7.3	26.9	45.6	14.5	0.8	148.3	997.5	33.0	287.8	436.6
4/6/2013	95.0±3.6	21.4	67.8	17.1	1.2	122.3	1004.3	23.0	15.3	28.1
7/6/2013	107.2±1.3	20.2	85.3	20.2	0.6	132.8	1001.1	21.3	21.6	47.8
11/6/2013	61.5±2.5	21.2	60.4	14.9	1.1	181.1	1000.9	25.6	219.3	387.6
14/6/2013	92.3±7.0	27.4	33.9	12.3	1.4	177.3	1000.4	32.3	255.3	433.3
17/7/2013	41.3±4.5	28.1	47.3	17.2	1.0	147.2	997.7	34.1	271.6	480.1
26/7/2013	46.8±2.1	25.9	68.1	22.6	0.9	174.8	995.9	28.1	46.9	95.9
27/7/2013	91.2±3.8	25.3	80.5	26.0	0.4	73.5	993.4	28.1	50.2	100.2
18/8/2013	21.7±12.9	30.4	32.7	13.9	1.6	224.1	1000.6	35.0	282.6	496.8
22/8/2013	128.8±2.7	24.7	74.8	23.1	0.4	232.3	1001.5	25.5	46.2	86.8
26/8/2013	40.0±2.7	26.6	52.6	17.7	0.9	147.5	998.4	29.4	226.6	381.9
28/8/2013	69.7±7.2	ND	ND	ND	ND	ND	ND	ND	ND	ND
1/9/2013	37.5±6.4	ND	ND	ND	ND	ND	ND	ND	ND	ND

Date		T_a	RH	P_{vapor}	WS	WD	P	T_g	R_{total}	PAR
9/9/2013	71.8±4.5	ND	ND	ND	ND	ND	ND	ND	ND	ND
13/9/2013	66.2±3.4	ND	ND	ND	ND	ND	ND	ND	ND	ND
29/9/2013	154.3±10.5	17.8	71.6	14.6	0.7	169.0	1003.0	18.2	22.1	37.3
4/10/2013	107.6±31.6	17.9	65.1	13.2	0.3	123.9	1006.8	20.1	135.3	193.3
12/10/2013	57.0±5.0	16.9	47.7	8.9	0.6	149.0	1010.8	18.6	114.5	170.6
16/10/2013	71.9±36.5	13.1	40.0	5.5	0.5	175.8	1018.0	15.2	162.8	216.4
17/10/2013	142.6±36.6	13.0	45.0	6.3	0.5	161.4	1018.3	14.7	141.1	186.4
26/10/2013	50.0±2.9	11.7	38.9	5.1	0.6	171.4	1015.0	12.9	126.0	161.4
2/11/2013	181.4±4.4	9.7	74.2	8.8	0.4	195.5	1014.6	11.0	77.0	101.8
5/11/2013	99.9±5.8	10.3	50.9	6.3	0.6	190.8	1013.0	11.3	54.8	77.1
7/11/2013	28.4±7.0	10.1	25.5	3.1	1.0	206.8	1013.4	10.4	100.3	136.0
8/11/2013	96.3±8.2	8.0	40.5	4.3	0.4	215.1	1012.8	8.8	70.1	90.8
11/11/2013	45.1±5.4	8.8	20.0	2.2	1.4	180.5	1020.8	9.5	85.8	122.0
13/11/2013	120.9±6.3	6.7	54.4	5.3	0.3	177.6	1013.5	7.6	45.2	60.9
22/11/2013	126.9±10.4	6.9	38.2	3.6	0.6	160.5	1016.7	7.1	60.1	82.6
30/11/2013	35.9±19.2	7.4	18.5	1.9	2.3	265.9	1015.2	7.9	83.6	101.4
1/12/2013	72.6±35.8	5.9	26.9	2.4	0.8	191.1	1015.4	6.5	75.6	90.5
6/12/2013	141.9±54.0	5.0	33.9	2.9	0.8	196.8	1012.0	5.9	65.2	79.6
7/12/2013	214.3±46.6	3.5	54.1	4.3	0.6	126.6	1012.3	4.2	50.9	60.7
22/12/2013	153.8±17.4	-1.5	33.4	1.8	0.8	163.7	1024.5	-0.8	59.2	103.2

[a] ND: no data was collected in this day. T_a: the daily average air temperature. RH: relative humidity. P_{vapor}: vapor pressure. WS: wind speed. WD: wind direction. P: atmospheric pressure. T_g: ground temperature. R_{total}: radiation. PAR: photosynthetically active radiation.

[b] Mean ±SD from 24 measurements in a day ($n = 24$).

Table S2. Concentrations of Si and chemical components indicative of secondary

aerosols (NH$_4^+$, NO$_3^-$, SO$_4^{2-}$, OC, and EC) in PM$_{2.5}$ samples in 2013.[a]

Date	Si (µg/m^3)[b]	SO$_4^{2-}$ (µg/m^3)	NO$_3^-$ (µg/m^3)	NH$_4^+$ (µg/m^3)	OC (µg/m^3)	EC (µg/m^3)
6/1/2013	0.90±0.05	22.20±0.89	16.41±0.82	15.81±0.63	53.32±2.67	10.30±0.51
11/1/2013	2.09±0.01	52.01±2.08	26.14±1.31	27.96±1.12	48.17±2.41	8.64±0.43
18/1/2013	0.64±0.02	39.27±1.57	31.00±1.55	26.75±1.07	61.79±3.09	16.61±0.83
25/1/2013	1.03±0.02	11.34±0.45	12.16±0.61	10.94±0.44	7.81±1.26	-
29/1/2013	1.30±0.04	112.87±4.51	69.30±3.47	62.01±2.48	39.70±1.99	10.63±0.53
2/2/2013	0.42±0.02	13.81±0.55	15.81±0.79	13.37±0.53	30.40±1.52	6.81±0.34
8/2/2013	0.70±0.03	30.69±1.23	7.56±0.38	6.59±0.26	18.49±0.92	1.80±0.09
11/2/2013	5.61±0.19	31.45±1.26	6.79±0.34	6.22±0.25	23.95±1.20	0.40±0.02
23/2/2013	0.99±0.03	43.09±1.72	16.83±0.84	13.11±0.52	27.24±1.36	3.22±0.16
24/2/2013	1.68±0.04	34.33±1.37	28.19±1.41	24.69±0.99	-	-
27/2/2013	1.48±0.05	53.59±2.14	30.76±1.54	23.57±0.94	40.57±2.03	4.13±0.21
2/3/2013	0.84±0.03	15.88±0.64	5.46±0.27	3.69±0.15	17.44±0.87	0.85±0.04
5/3/2013	1.11±0.01	16.28±0.65	47.42±2.37	25.53±1.02	38.70±1.94	16.78±0.84
6/3/2013	1.38±0.05	30.27±1.21	64.44±3.22	32.22±1.29	37.54±1.88	14.12±0.71
7/3/2013	12.92±0.45	33.92±1.36	68.69±3.43	34.65±1.39	41.69±2.08	18.11±0.91
10/3/2013	0.87±0.01	9.62±0.38	14.59±0.73	11.55±0.46	16.78±0.84	6.48±0.32
14/3/2013	0.31±0.02	30.91±1.24	37.69±1.88	24.32±0.97	19.93±1.00	6.81±0.34
15/3/2013	0.48±0.02	57.06±2.28	79.64±3.98	45.59±1.82	22.43±1.12	10.30±0.51
26/3/2013	0.71±0.02	45.55±1.82	48.63±2.43	29.79±1.19	17.11±0.86	6.15±0.31
31/3/2013	0.83±0.02	56.52±2.26	42.55±2.13	31.61±1.26	14.62±0.73	5.48±0.27
2/4/2013	0.88±0.02	21.91±0.88	9.26±0.46	14.58±0.58	13.14±0.66	2.08±0.10
12/4/2013	0.28±0.01	4.39±0.18	6.02±0.30	3.53±0.14	10.89±0.54	3.03±0.15
21/4/2013	0.48±0.01	22.03±0.88	32.01±1.60	14.89±0.60	14.11±0.71	3.41±0.17
22/4/2013	1.17±0.03	35.63±1.43	45.61±2.28	22.55±0.90	16.86±0.84	3.65±0.18
6/5/2013	0.49±0.01	48.02±1.92	28.49±1.42	25.04±1.00	21.59±1.08	6.34±0.32
7/5/2013	0.95±0.02	47.85±1.91	23.58±1.18	25.22±1.01	23.63±1.18	7.62±0.38
17/5/2013	1.85±0.03	18.42±0.74	21.77±1.09	12.05±0.48	12.07±0.60	3.08±0.15
18/5/2013	1.34±0.02	25.90±1.04	18.50±0.93	13.08±0.52	10.65±0.53	3.65±0.18
21/5/2013	1.02±0.03	18.76±0.75	8.78±0.44	7.31±0.29	19.03±0.95	3.69±0.18
23/5/2013	3.85±0.07	11.10±0.44	6.02±0.30	4.73±0.19	12.41±0.62	3.03±0.15
3/6/2013	0.89±0.05	25.99±1.04	12.99±0.65	14.46±0.58	18.23±0.91	5.45±0.27

4/6/2013	1.62±0.05	23.84±0.95	21.77±1.09	17.30±0.69	14.02±0.70	5.21±0.26
7/6/2013	1.60±0.14	34.51±1.38	19.19±0.96	23.49±0.94	11.60±0.58	5.16±0.26
11/6/2013	0.66±0.03	14.68±0.59	15.19±0.76	5.99±0.24	13.33±0.67	2.70±0.14
14/6/2013	0.42±0.02	34.01±1.36	9.72±0.49	10.29±0.41	20.42±1.02	3.36±0.17
17/7/2013	0.78±0.04	7.49±0.30	3.60±0.18	3.24±0.13	12.17±0.61	1.91±0.10
26/7/2013	0.82±0.05	12.87±0.51	7.14±0.36	5.57±0.22	11.29±0.56	2.29±0.11
27/7/2013	1.44±0.02	21.09±0.84	23.23±1.16	13.51±0.54	16.78±0.84	3.25±0.16
18/8/2013	0.46±0.03	0.88±0.04	0.36±0.02	0.53±0.02	4.10±0.21	1.70±0.09
22/8/2013	0.26±0.01	20.34±0.81	11.43±0.57	12.39±0.50	17.80±0.89	10.10±0.51
26/8/2013	0.35±0.02	11.72±0.47	3.61±0.18	4.40±0.18	9.52±0.48	1.52±0.08
28/8/2013	0.48±0.02	17.47±0.70	15.92±0.80	10.44±0.42	13.85±0.69	1.48±0.07
1/9/2013	0.52±0.05	7.97±0.32	4.71±0.24	3.48±0.14	8.21±0.41	1.82±0.09
9/9/2013	1.01±0.04	27.85±1.11	12.16±0.61	11.21±0.45	9.87±0.49	1.20±0.06
13/9/2013	1.74±0.07	22.22±0.89	12.59±0.63	10.77±0.43	10.07±0.50	1.49±0.07
29/9/2013	0.89±0.02	44.85±1.79	38.65±1.93	21.69±0.87	24.58±1.23	3.13±0.16
4/10/2013	0.50±0.02	28.76±1.15	21.58±1.08	11.08±0.44	-	-
12/10/2013	2.30±0.09	9.05±0.36	10.17±0.51	4.07±0.16	12.93±0.65	2.64±0.13
16/10/2013	3.32±0.14	6.73±0.27	8.66±0.43	4.77±0.19	9.82±0.49	1.680±0.08
17/10/2013	0.48±0.01	54.13±2.17	27.74±1.39	24.45±0.98	15.48±0.77	3.16±0.16
26/10/2013	0.46±0.03	15.86±0.63	8.17±0.41	5.85±0.23	10.12±0.51	2.41±0.12
2/11/2013	0.72±0.02	49.28±1.97	48.17±2.41	27.83±1.11	25.18±1.26	6.93±0.35
5/11/2013	0.39±0.01	16.41±0.66	18.51±0.93	11.19±0.45	28.53±1.43	7.59±0.38
7/11/2013	1.54±0.08	5.39±0.22	3.03±0.15	2.13±0.09	10.77±0.54	1.26±0.06
8/11/2013	4.03±0.14	28.60±1.14	13.403±0.67	9.63±0.39	23.68±1.18	5.16±0.26
11/11/2013	0.41±0.01	12.43±0.50	3.70±0.19	2.19±0.09	9.42±0.47	1.36±0.07
13/11/2013	0.96±0.04	42.57±1.70	15.96±0.80	13.16±0.53	24.65±1.23	6.13±0.31
22/11/2013	1.86±0.09	42.59±1.70	19.21±0.96	11.56±0.46	27.83±1.39	5.80±0.29
30/11/2013	0.21±0.01	15.29±0.61	2.44±0.12	1.98±0.08	9.30±0.47	0.78±0.04
1/12/2013	1.23±0.06	24.42±0.98	6.41±0.32	4.70±0.19	20.74±1.04	3.55±0.18
6/12/2013	1.58±0.04	12.41±0.50	21.17±1.06	11.29±0.45	55.41±2.77	12.05±0.60
7/12/2013	3.35±0.09	39.42±1.58	35.78±1.79	26.14±1.05	62.07±3.10	12.78±0.64
22/12/2013	0.94±0.04	19.66±0.79	26.22±1.31	12.43±0.50	53.44±2.67	12.01±0.60

[a] In the measurement of secondary species, the uncertainties were 5%, 5%, 4%, 4%, and 5% for OC, EC, SO_4^{2-}, NH_4^+, and NO_3^-, respectively. See the Sect. 1.3 and 1.5, Supplement.

[b] Mean ±SD from three parallel measurement by ICP-MS ($n = 3$).

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